Plutonium: An anthropogenic element under nuclear forensic investigations

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Plutonium is an anthropogenic, i.e. man-made element.

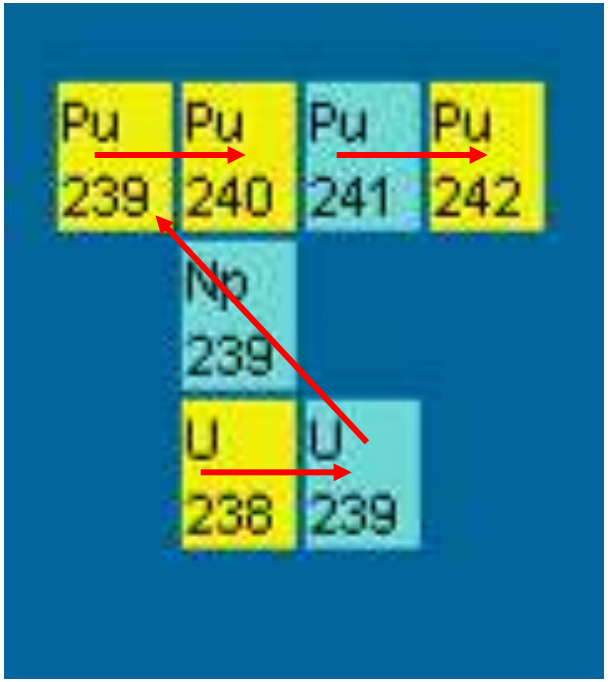
At the end of 1940 Pu-238 was first time synthesized:

U-238 (d,2n) Np-238 $\xrightarrow{\beta-}$ Pu-238

In nuclear reactor (~1% Pu in spent fuel):

U-238 + n $\rightarrow$ U-239 $\xrightarrow{2\ \beta-}$ **Pu-239** + n $\rightarrow$ **Pu-240** + n $\rightarrow$ **Pu-241** + n $\rightarrow$ **Pu-242**

U-235 + 2n $\xrightarrow{\beta-}$ U-237 $\xrightarrow{\beta-}$ Np-237 + n $\rightarrow$ Np-238 $\rightarrow$ **Pu-238**



Main questions to be answered:

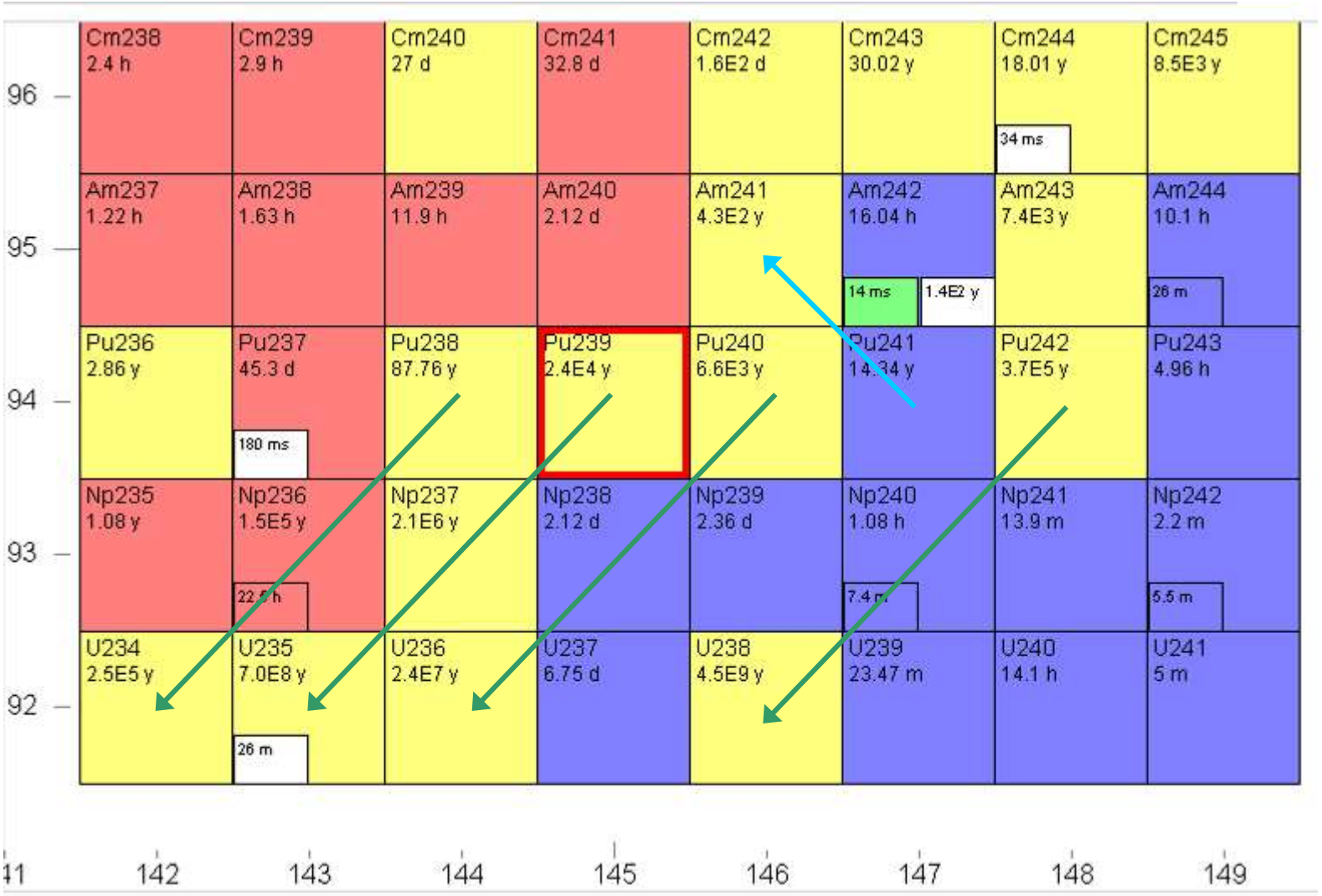
1. **Intended use** of the material
   (nuclear weapons, fuel, scrap,…)

2. **Origin**
   (place of production, process, place of diversion, last legal owner,…)

Answers:

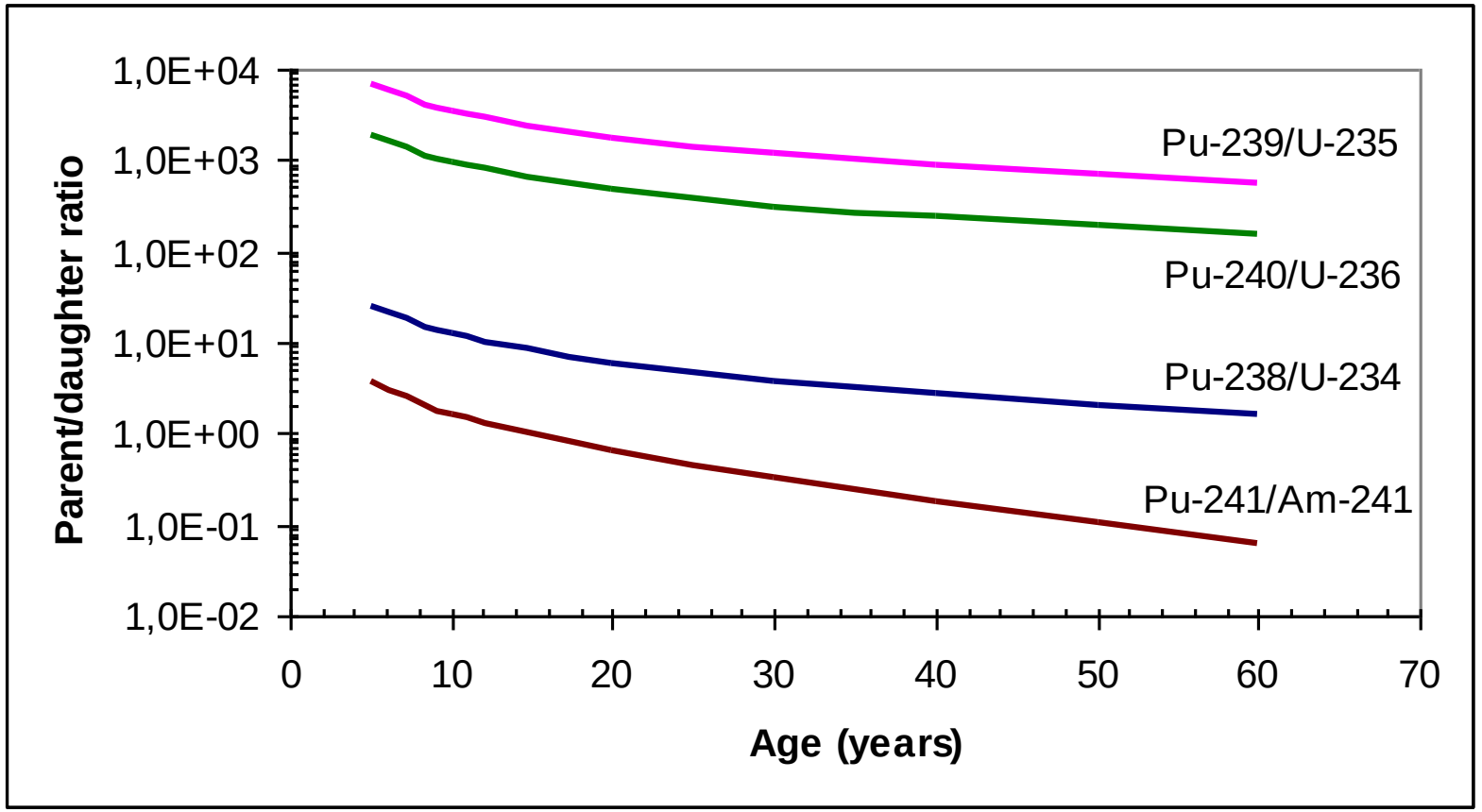
- isotopics
- Pu-content
- age
- impurities
- microstructure
- …

Plutonium decay



Age determination

Age = the time elapsed since the last chemical processing (e.g. production, reprocessing, purification)



Age determination - limitations

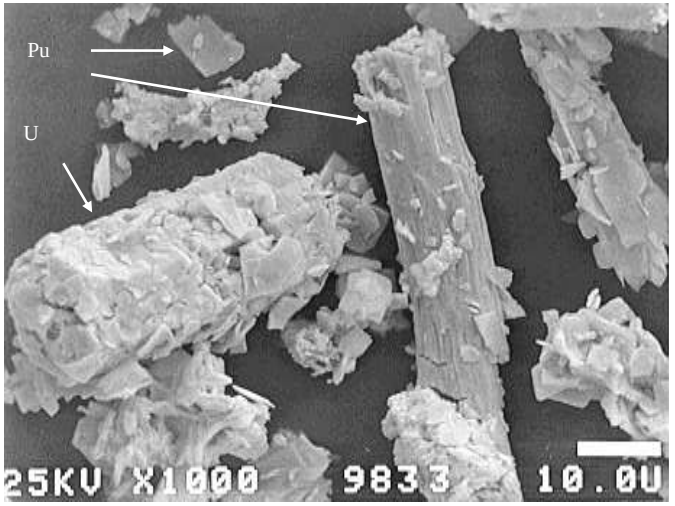
- Incomplete Pu/U/Am separation when reprocessing

  ⇒ U and Am remainings

  ⇒ inconsistent results

Sample	$^{241}Pu/^{241}Am^{*}$	$^{238}Pu/^{234}U$	$^{239}Pu/^{235}U$	$^{240}Pu/^{236}U$
RR	6.1	**2.35** ± 0.10	**2.66** ± 0.26	**2.29** ± 0.05
F19	20.6	**1.18** ± 0.07	**51.9** ± 3.8	**19.9** ± 0.3

Age by SIMS in February 2000. * By gamma spectrometry.

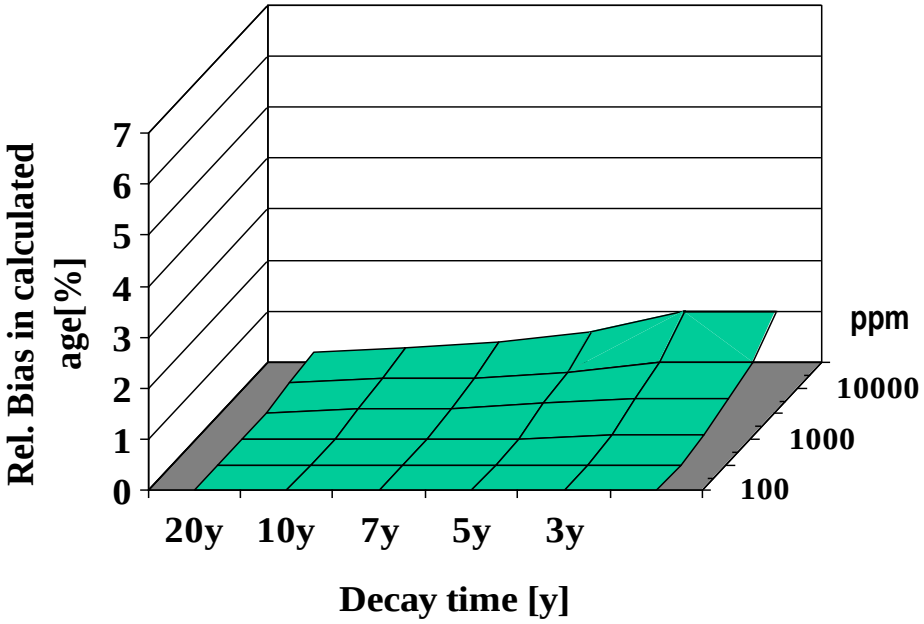


- MOX

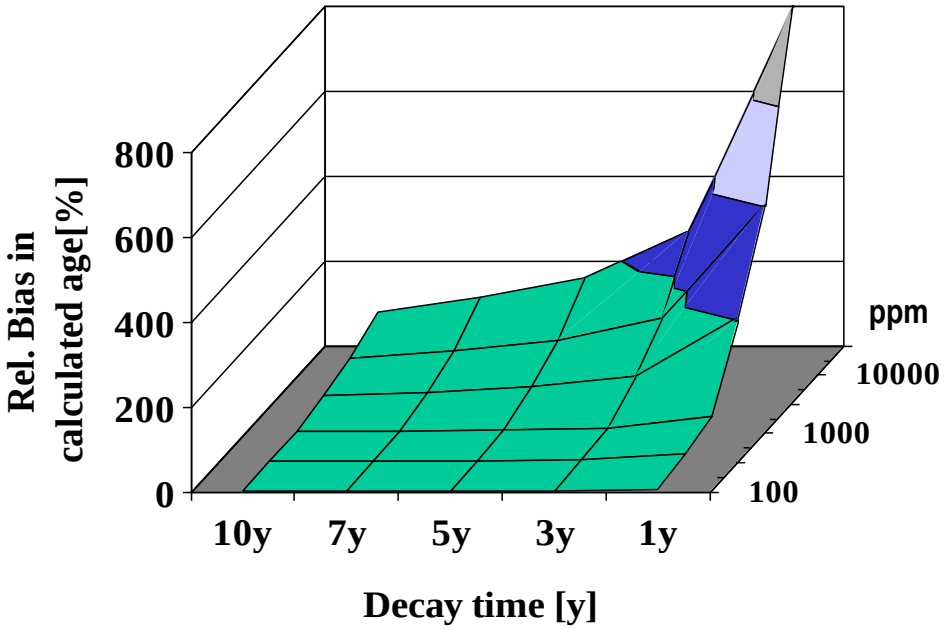
  ⇒ U interference

Effect of the residual U on the age

^{238}Pu / ^{234}U
reactor grade



^{238}Pu / ^{234}U
weapons grade



Plutonium composition

Weapons grade:

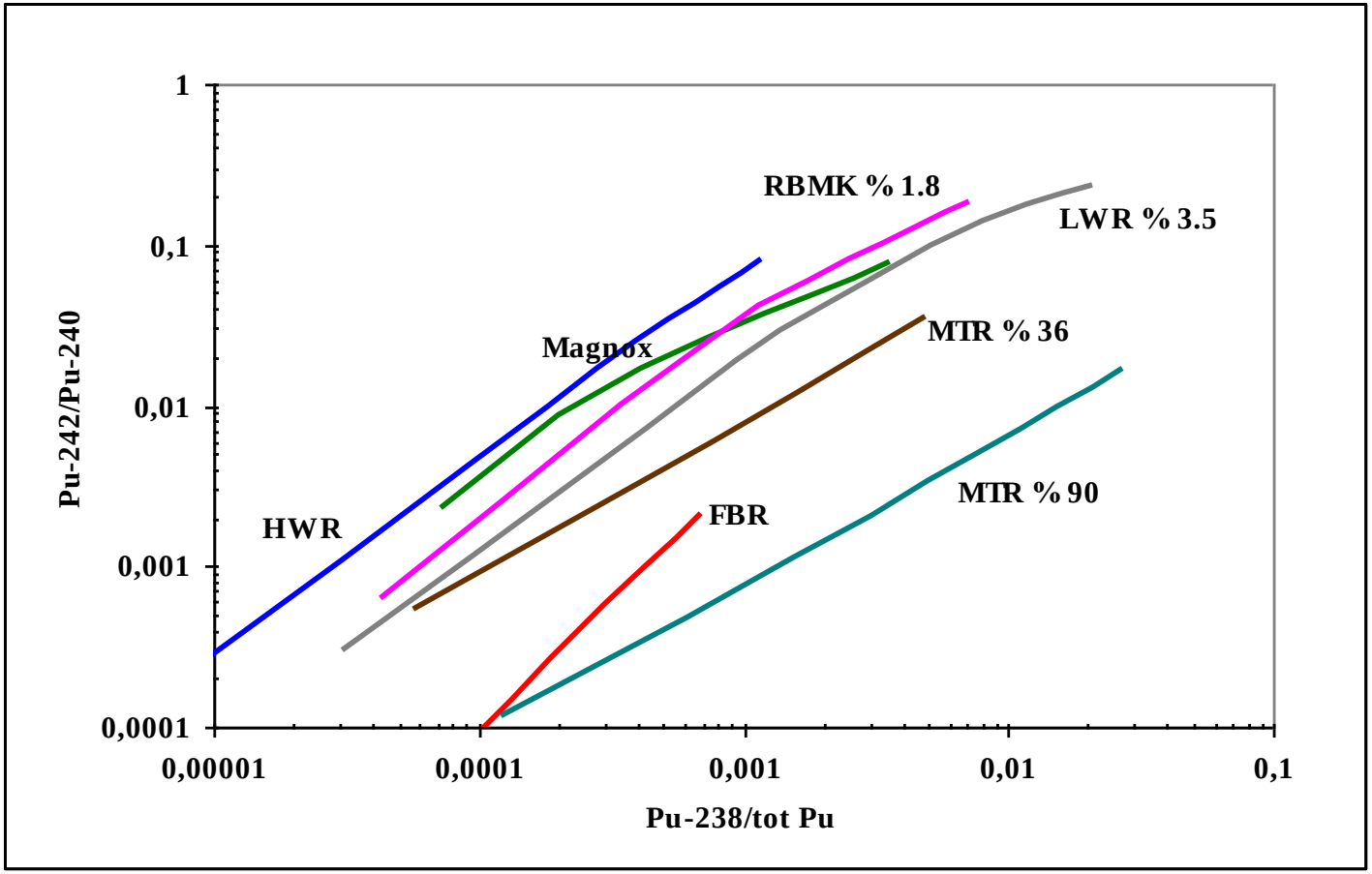
- Pu-240 < 7 %
- 240/239 < 0.07

Reactor grade:

- Pu-239 = 45-60%
- Pu-240 = 20-35 %
- 240/239 = 0.3-0.8

The isotopic composition of the reactor produced plutonium is depending on the initial U-235 enrichment, neutron spectrum (soft >< hard), flux and irradiation time.

Reactor type determination

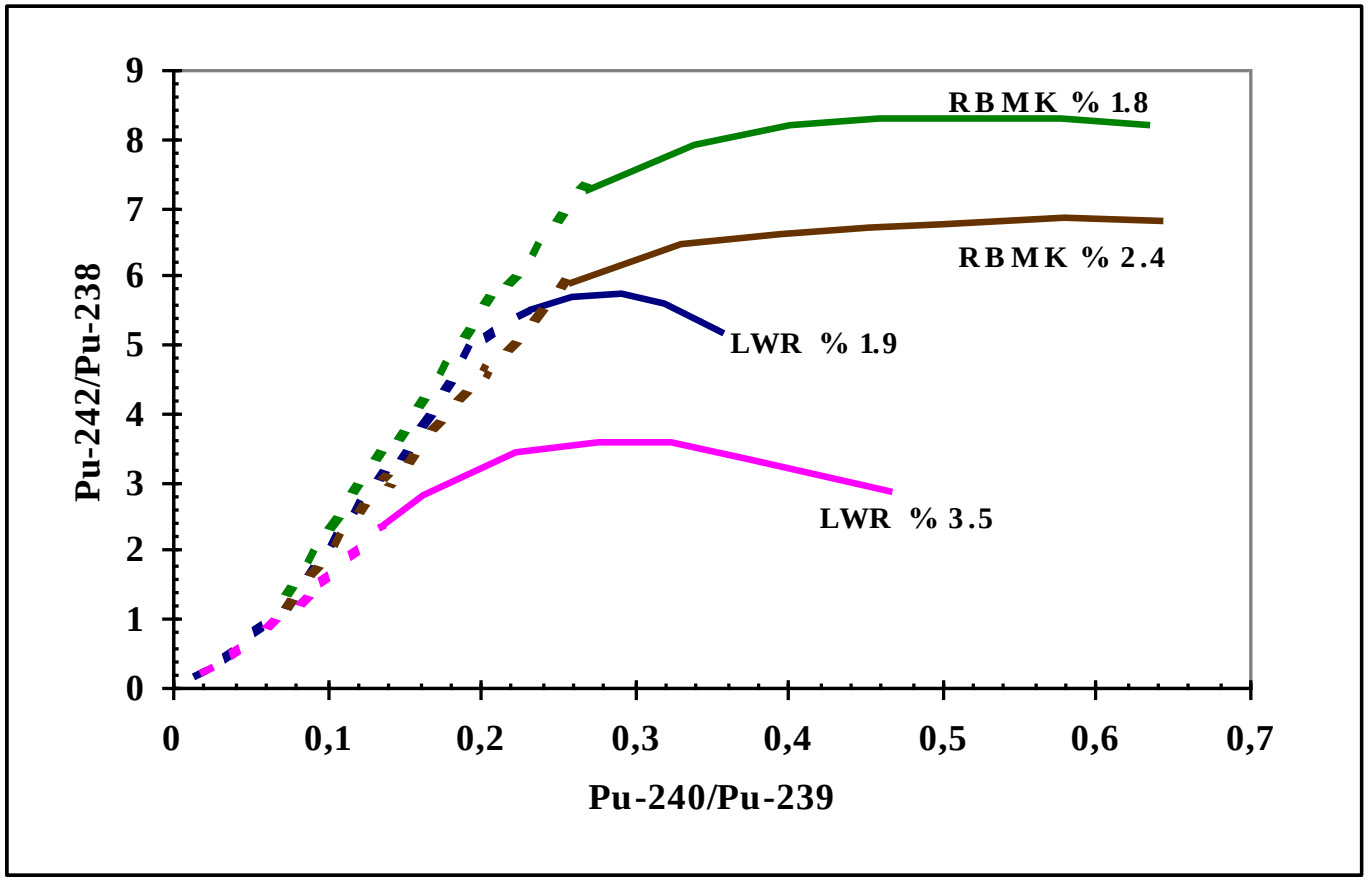


Softer the neutron spectrum – higher the $^{242}Pu/^{240}Pu$ ratio

Higher the initial ^{235}U enrichment – higher the ^{238}Pu abundance

Reactor type determination

Difference between light-water and graphite-moderated reactors.



Case studies

Find 13- Konstanz

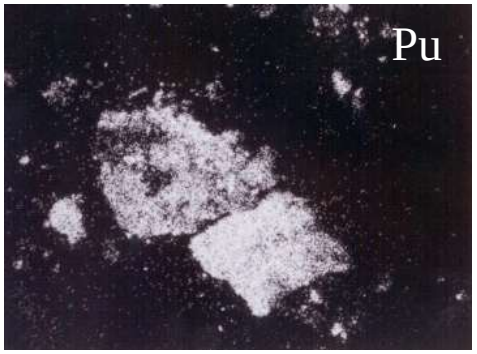
56 g of powder containing Pu

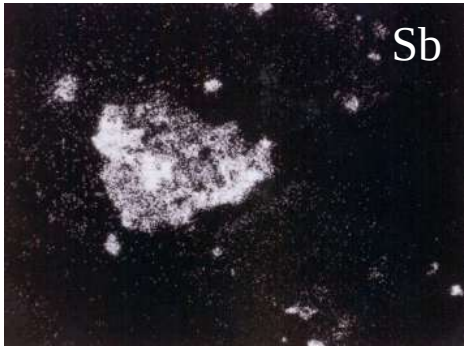
Isotope/ element	w-%
^{238}Pu	0.067
^{239}Pu	99.751
^{240}Pu	0.181
Mercury	61
Antimony	11
Oxygen	6
Plutonium	10
Gallium	0.15 (4.6 a-%)
Iodine	1.6

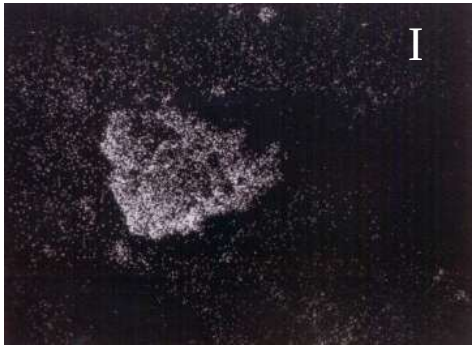
$Hg_2Sb_2O_7$ = "Red mercury" !

Find 13- Konstanz

Pu particle and powder agglomerate - electron probe







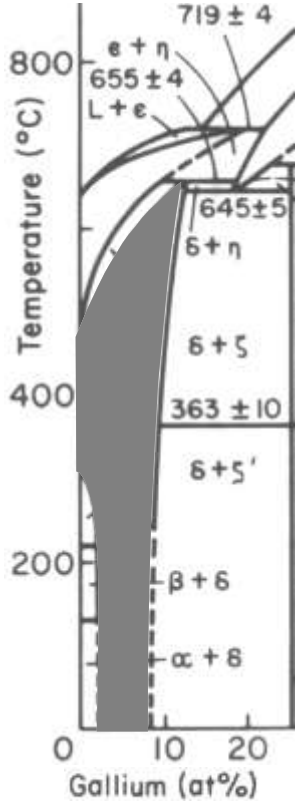
Find 13- Konstanz

“Red mercury”:

- Anti-Radar-Shielding for aircraft’s
- self-guided warheads
- nuclear bombs smaller and more efficient
- density 23 g/ccm

Plutonium:

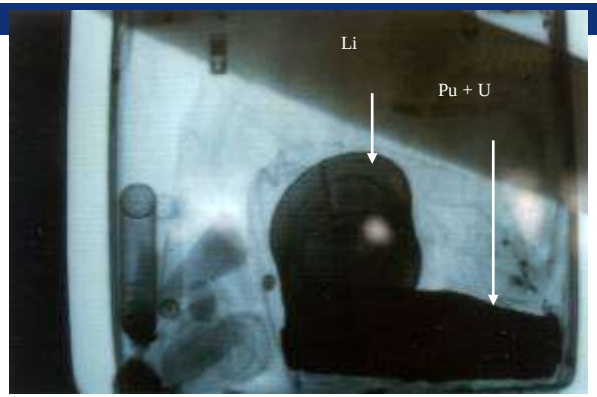
- weapons-grade Pu
- Ga is used to stabilise the δ- phase of Pu

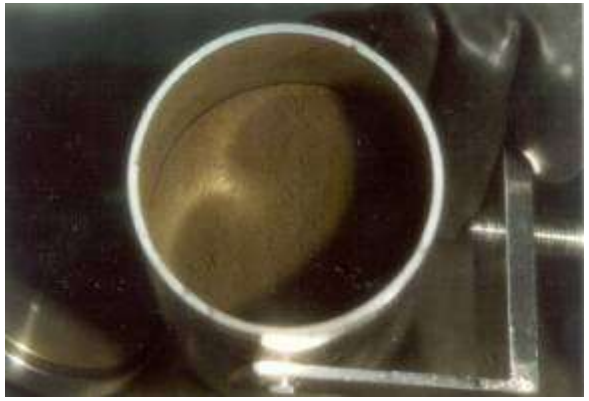


Compound with these characteristics has never been found !!

Example of the proliferation of classified knowledge !

Find 19 - Munich





Mixed plutonium and uranium oxides:

363 g Pu - 121 g U

Pu	238	239	240	241	242
w-%	0.17	87.58	10.78	0.81	0.66

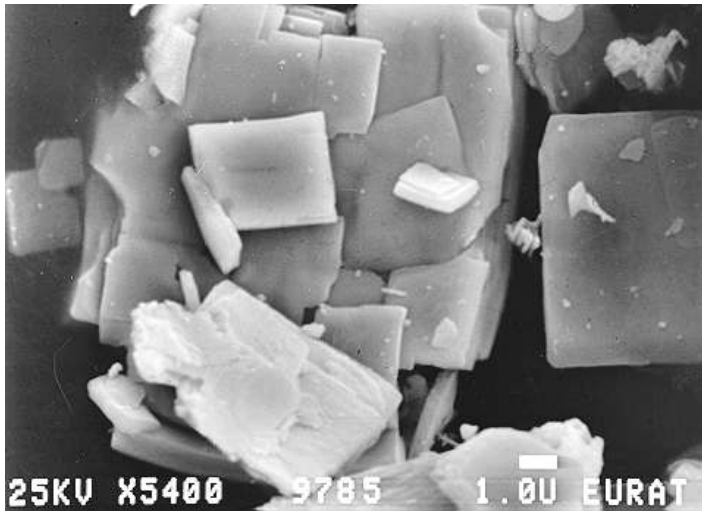
U	234	235	236	238
w-%	0.020	1.60	0.048	98.35

201 g of Li-metal enriched to 89.4 % in ^{6}Li.

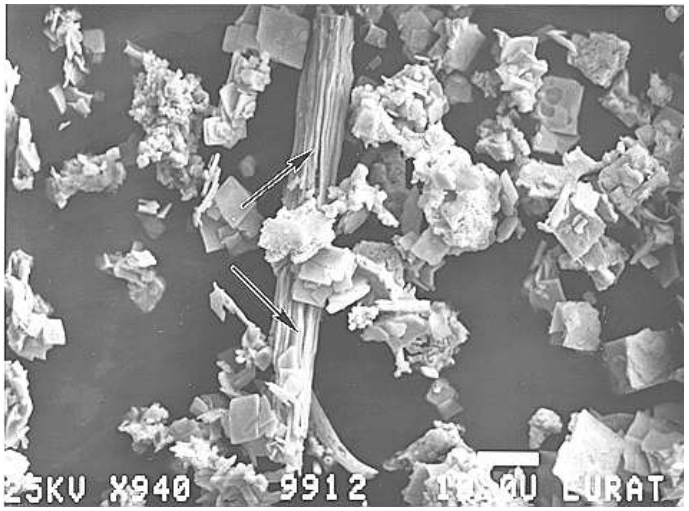
Find 19 - Munich

Powder consists of two different Pu particle types:

1) platelets of PuO_2



2) rod-shaped PuO_2

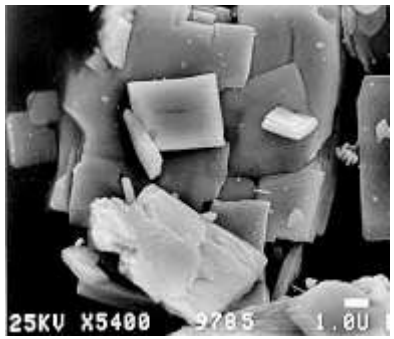


	Bulk	Particle 1	Particle 2
$^{240}Pu/^{239}Pu$	0.1226	0.1159	0.1245
Age from the $^{240}Pu/^{236}U^{*}$	(20.6)**	21.5	19.9

*Age by SIMS in February 2000. ** By gamma spectrometry from the Am-241 build-up.

Find 19 - Munich

Microstructural fingerprint



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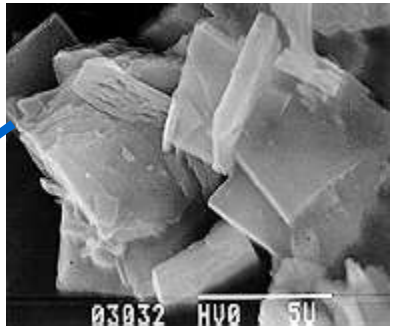
SEM



Reference SM3-1

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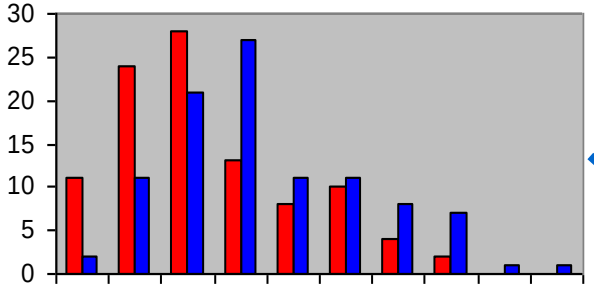
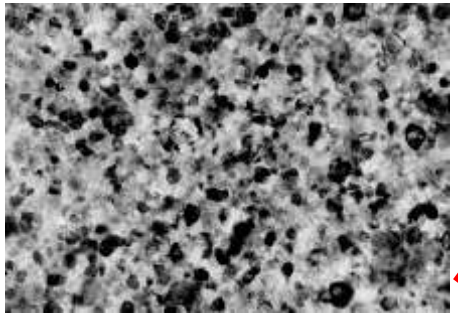
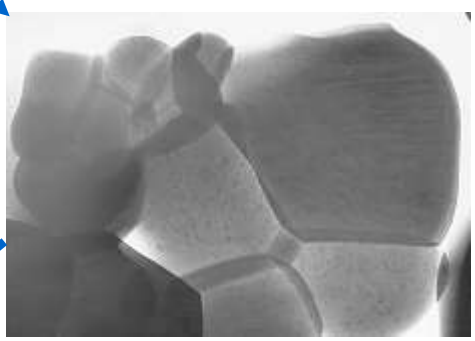




TEM



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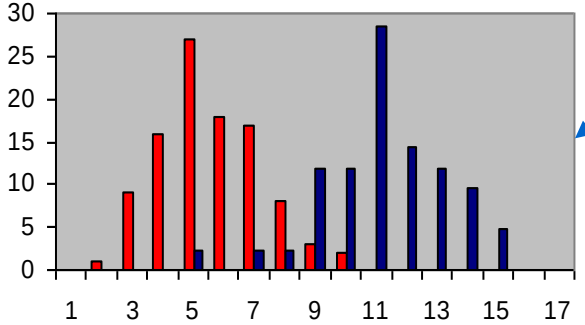


Find 19 - Munich

Plutonium

- close to weapons-grade
- not from commercial reprocessing or from military production
- mixture of residues of different types of spent fuels

Lithium-6

- energetic tritons via $^{6}Li(n,\alpha)^{3}T$
- tritium can initiate DT nuclear fusion in a thermonuclear weapon

May not be a coincidence to find such materials together !!

WAK – Reprocessing plant (decommissioned)

Contamination of an employee was detected in a routine urine control.

Find 24 - Karlsruhe

"Players":

Mr. M. – The thief

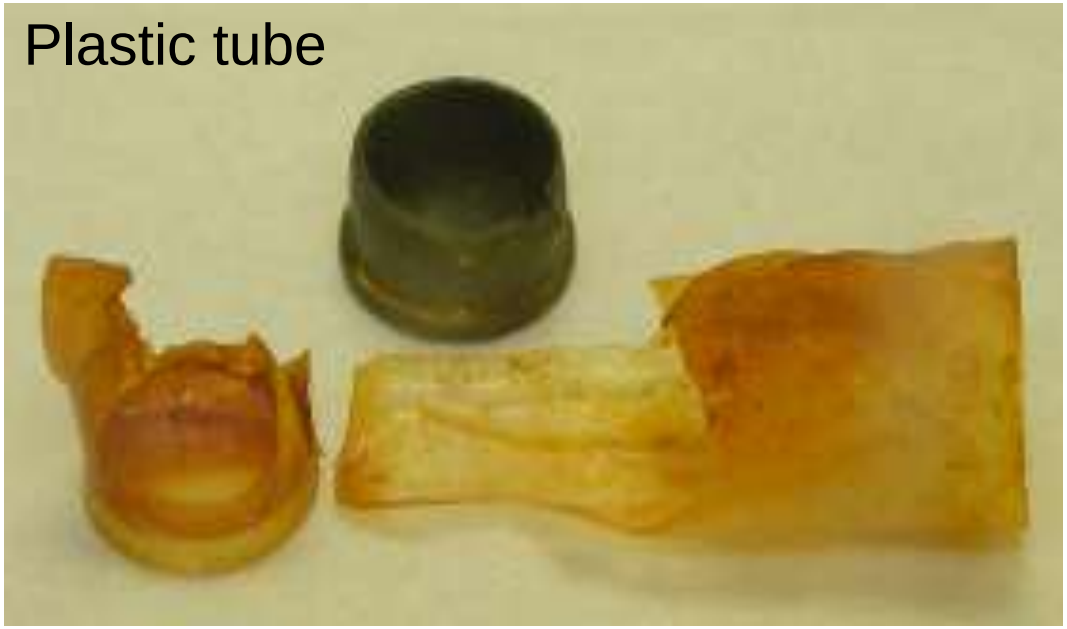
Mrs. E. - Girl friend of Mr. M.

Ms. A. E. - Daughter of Ms. E.

Find 24 - Karlsruhe

Stolen items:

Plastic tube



Swipe cloth

Total of a few milligrams of radioactive material including Pu, U, ^{241}Am, $^{134+137}$Cs, ^{154}Eu and ^{125}Sb.

Find 24 - Karlsruhe

Estimated (ICRP Model) activity uptake (in kBq):

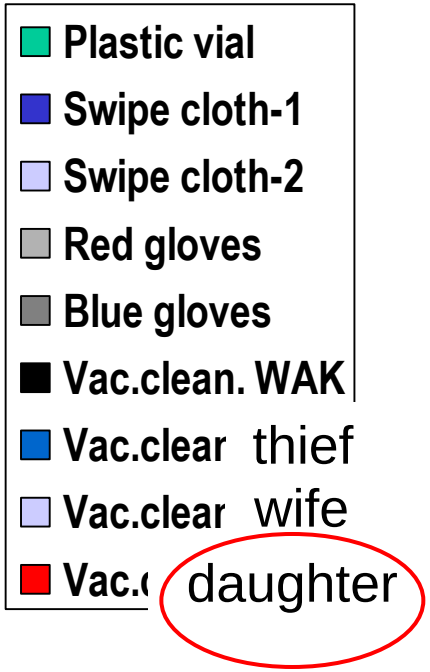
Person	Way of uptake	Cs-137	Am-241	Pu-241
Mr. M.	Inhalation		4	16
	Ingestion		-	-
Mrs. E.	Inhalation		28	1110
	Ingestion	118	3450	13800
Ms. A.E.	Inhalation		0,4	
	Ingestion	3	69	

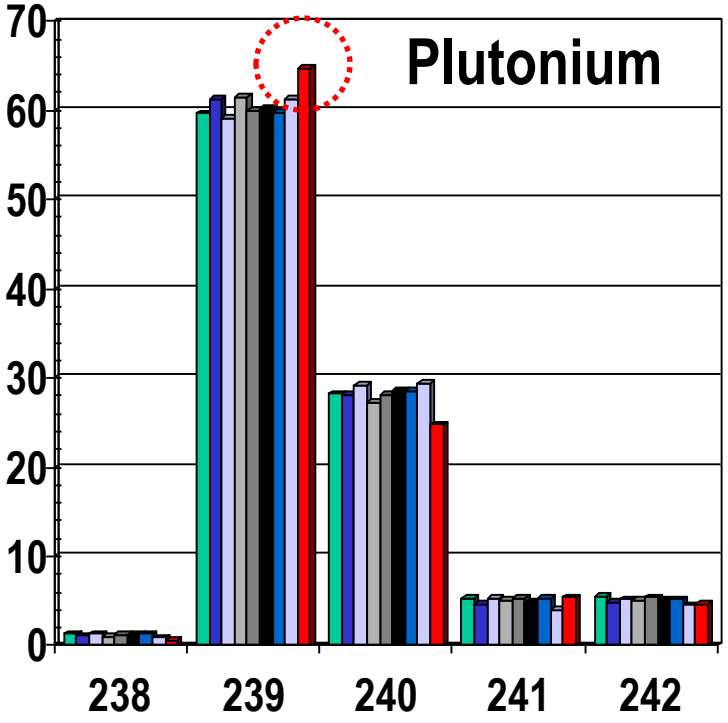
Find 24 - Karlsruhe

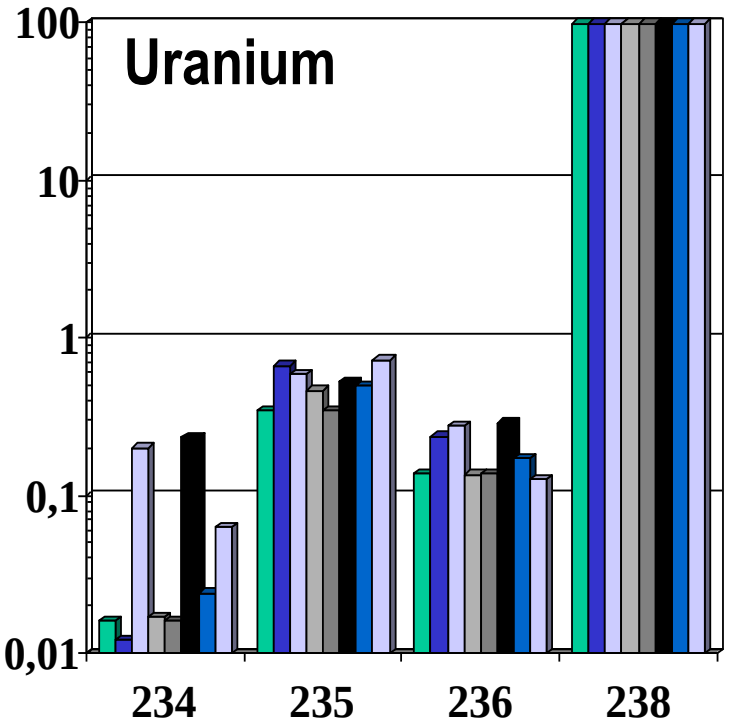
Can the contaminations and incorporations be explained by the two sources, vial + cloth ?

- Analyse samples from different locations
  - three apartments (thief, girl friend, daughter's boyfriend)
  - facility
- Compare
  - samples
  - external information (which fuel reprocessed, WAK, …)

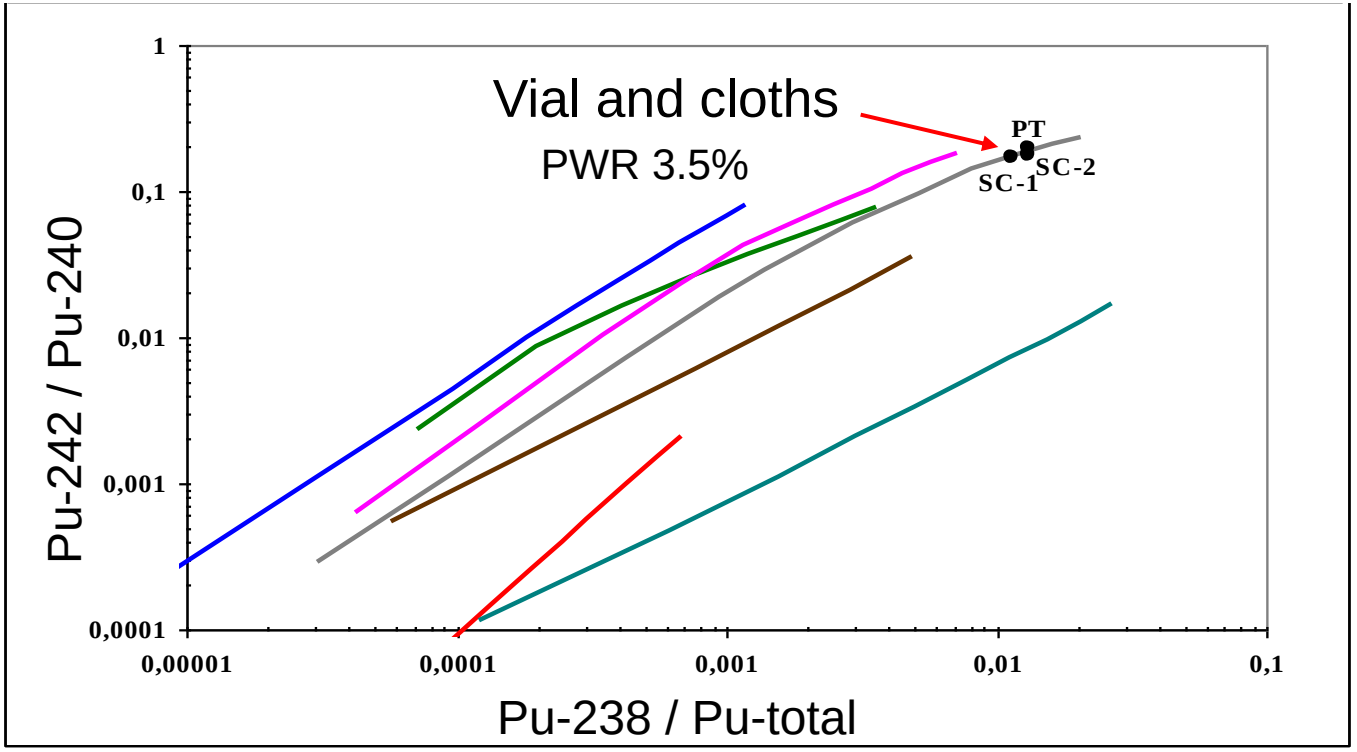
Isotopic compositions:

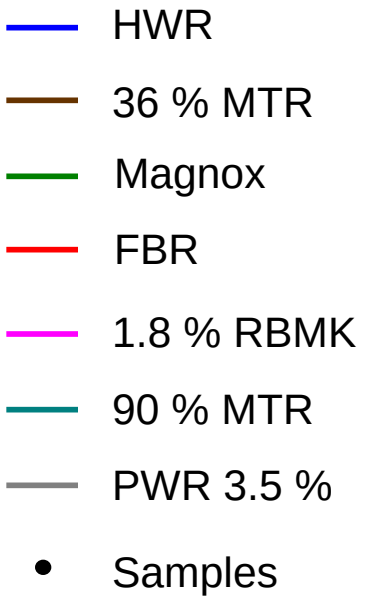






Find 24 - Karlsruhe





– Pu composition matched with the reprocessed LWR fuel

– The age of the Pu was 12-14 years, which was the time of the last reprocessing campaign

Find 24 - Karlsruhe

- Vacuum cleaner bag from “the daughter” showed significantly different Pu isotopic composition

  ⇒ Pu-240/Pu-239 = 0.38 rather than 0.47

  ⇒ mixture of reactor Pu (~ 90%) and fallout Pu (~10%)

- The plastic vial can be excluded as source of incorporation of the wife (^{137}Cs content too low)
  Swipe cloth:
  Varying elemental ratios can be explained by different solubilities (Cs-137 dissolves well in water, Pu and Am not)

- Decontamination of two apartments ~ 2 M€

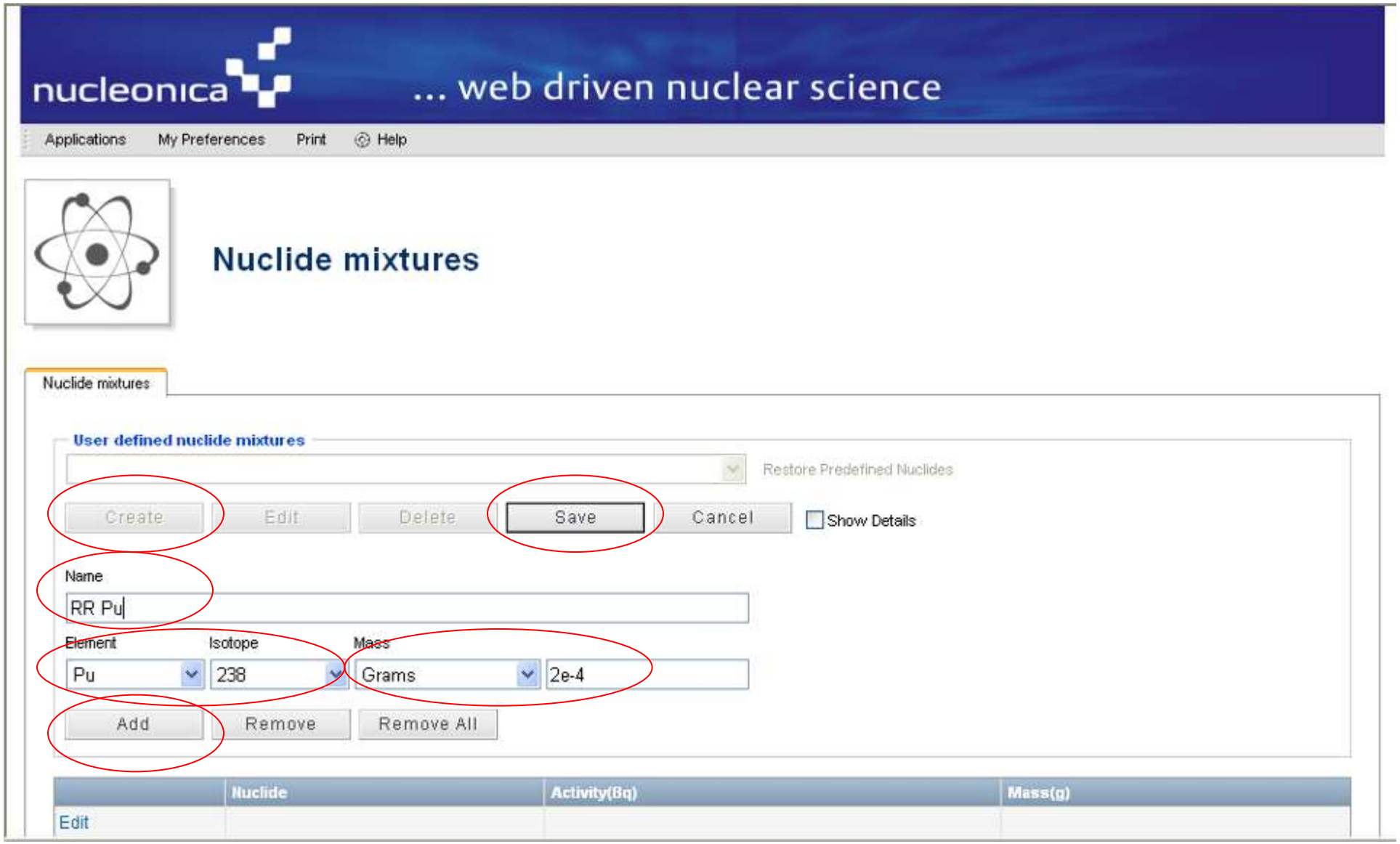
Exercises

Exercises

1. Which parent/daughter and parent/grand-daughter ratios can be used to determine the age of a 10 mg Pu-sample with the following composition (wt-%):

   Pu-238: 2 %
   Pu-239: 57 %
   Pu-240: 25 %
   Pu-241: 10 %
   Pu-242: 6 %

   Remarks: Method of the choice is ICP-MS without chemical separation with detection range of 10^8 (i.e. the parent/daughter ratio cannot be larger than 10^8).


nucleonica
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Applications
My Preferences
Print
Help
Nuclide mixtures
Nuclide mixtures
User defined nuclide mixtures
Restore Predefined Nuclides
Create
Edit
Delete
Save
Cancel
Show Details
Name
RR Pu
Element
Isotope
Mass
Pu
238
Grams
2e-4
Add
Remove
Remove All
Nuclide
Activity(Bq)
Mass(g)
Edit

Exercise 1

Nuclide mixtures

User defined nuclide mixtures

RR Pu — Restore Predefined Nuclides

Create | Edit | Delete | Save | Cancel | Show Details

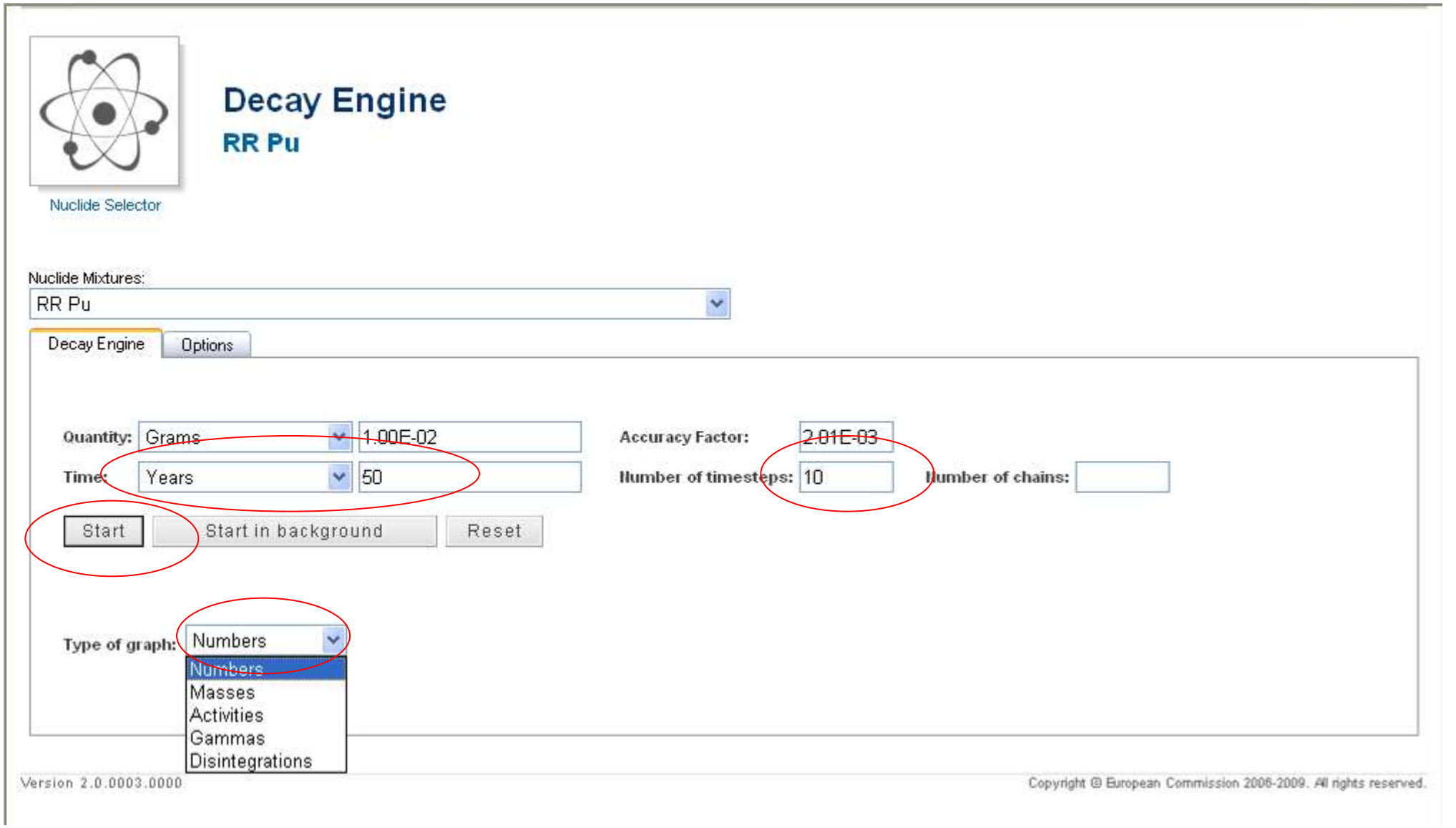
Name: RR Pu

Element: Ac | Isotope: 206 | Mass: Grams | 1

Add | Remove | Remove All

	Nuclide	Activity(Bq)	Mass(g)
Edit	94 Pu238	1.2672E+8	2.0000E-4
Edit	94 Pu239	1.3080E+7	5.7000E-3
Edit	94 Pu240	2.0990E+7	2.5000E-3
Edit	94 Pu241	3.8292E+9	1.0000E-3
Edit	94 Pu242	8.7783E+4	6.0000E-4
Edit			
Edit			
Edit			

Decay Engine | Gamma Dosimetry & Shielding


Decay Engine
RR Pu
Nuclide Selector
Nuclide Mixtures:
RR Pu
Decay Engine
Options
Quantity:
Grams
1.00E-02
Accuracy Factor:
2.01E-03
Time:
Years
50
Number of timesteps:
10
Number of chains:
Start
Start in background
Reset
Type of graph:
Numbers
Numbers
Masses
Activities
Gammas
Disintegrations
Version 2.0.0003.0000
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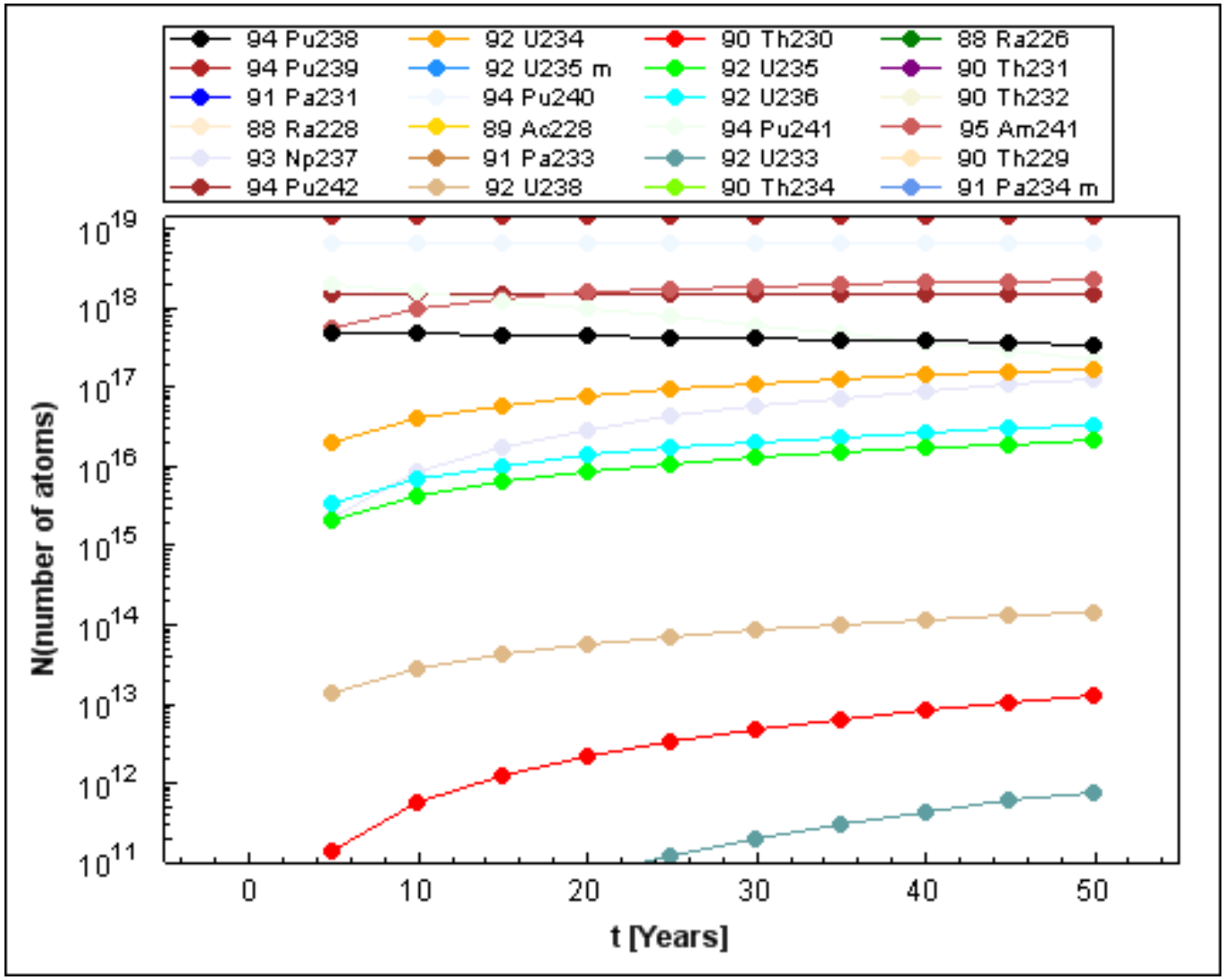
Quantity: Grams 1.00E-02 Accuracy Factor: 2.01E-03

Time: Years 50 Number of timesteps: 10 Number of chains: 8

Start | Start in background | Reset | Show details | Create Nuclide Mixture

Parent+Daughters	Half-life	N(atoms)	M(g)	A(Bq)	G(keV/s)
94 Pu238	87.76 y	3.41E+17	1.35E-04	8.54E+07	2.24E+06
92 U234	2.5E5 y	1.65E+17	6.41E-05	1.48E+04	1.58E+03
90 Th230	7.5E4 y	1.24E+13	4.73E-09	3.61E+00	1.34E+00
88 Ra226	1.6E3 y	1.95E+09	7.31E-13	2.67E-02	1.76E-01
86 Rn222	3.82 d	1.27E+04	4.69E-18	2.67E-02	1.04E-02
84 Po218	3.1 m	7.17E+00	2.59E-21	2.67E-02	2.46E-04
82 Pb214	26.8 m	6.20E+01	2.20E-20	2.67E-02	5.84E+00
83 Bi214	19.9 m	4.60E+01	1.64E-20	2.67E-02	4.11E+01
84 Po214	1.6E2 µs	0	0	0	0
82 Pb210	22.17 y	8.06E+06	2.81E-15	7.99E-03	1.57E-02
83 Bi210	5.01 d	4.99E+03	1.74E-18	7.98E-03	2.34E-06
84 Po210	1.4E2 d	1.32E+05	4.61E-17	7.67E-03	7.45E-05
82 Pb206 Stable	stable	2.54E+06	8.68E-16	0	0
94 Pu239	2.4E4 y	1.43E+19	5.69E-03	1.31E+07	8.35E+05
92 U235 m	26 m	2.94E+10	1.15E-11	1.31E+07	2.51E-06
92 U235	7.0E8 y	2.06E+16	8.04E-06	6.43E-01	9.55E+01
90 Th231	1.06 d	8.52E+04	3.27E-17	6.43E-01	8.46E+00
91 Pa231	3.3E4 y	5.07E+08	1.94E-13	3.40E-04	1.07E-02

Exercise 1


94 Pu238
94 Pu239
91 Pa231
88 Ra228
93 Np237
94 Pu242
92 U234
92 U235 m
94 Pu240
89 Ac228
91 Pa233
92 U238
90 Th230
92 U235
92 U236
94 Pu241
92 U233
90 Th234
88 Ra226
90 Th231
90 Th232
95 Am241
90 Th229
91 Pa234 m
N(number of atoms)
10^{19}
10^{18}
10^{17}
10^{16}
10^{15}
10^{14}
10^{13}
10^{12}
10^{11}
0
10
20
30
40
50
t [Years]

Exercises

2. In which of the following reactors was the Pu from previous example most likely produced ?

- BWR, 3.3 % U-235, 40 Burn-up, 8 years decay
- PWR, 3.3 % U-235, 40 Burn-up, 8 years decay
- BWR, 1.9 % U-235, 22 Burn-up, 8 years decay

Remember: Pu-240/Pu-239 = 0.44
Pu-241/Pu-239 = 0.18

webKORIGEN

webKORIGEN was developed from the Oak Ridge Isotope Generation and Depletion code ORIGEN.
Starting with a given initial reactor fuel or a single target nuclide, it calculates the time evolution of nuclide densities changing due to decays and neutron-induced reactions, and determines derived nuclear properties such as masses, radioactivities, heat releases, radiotoxicities, emission of radiation, etc...

Step 1: Calculation Mode | Step 2: Reactor / Operation | Step 3: Input Summary and Run | Step 4: Display Results | Step 5: Log files

- ○ Reactor irradiation
  - Power
  - Flux
- ○ Decay
- ◉ Reactor irradiation and decay
- ○ Reactor irradiation, cooling, reprocessing and decay of waste

Step 1: Calculation Mode | Step 2: Reactor / Operation | Step 3: Input Summary and Run | Step 4: Display Results | Step 5: Log files

Reactor type

- PWR
- BWR
- EFR

Reactor Parameters

Burnup (MW_{th} d / kg_{IHM}): 40

Total initial heavy metal mass (t_{IHM}): 20

Electrical efficiency (%): 34

Derived Power values

Specific Power:

Thermal Power:

Electrical Power:

Update power values

Neutron Spectrum

- UOX
  - U235/U (w/o): 3.3
- MOX

Nuclide	Weight (%)
Pu238/Pu (w/o)	2.6
Pu239/Pu (w/o)	50.5
Pu240/Pu (w/o)	27.8
Pu241/Pu (w/o)	11.5
Pu242/Pu (w/o)	7.6
Am241/Pu (w/o)	1.0

Uranium matrix

- Natural
- Depleted

Pu_{fiss}/(U+Pu) (w/o): 3.7

Irradiation and decay parameters

No. of cycles	5	
Length of cycle	365.24	d
Load factor (%)	80.0	
Fuel decay time after discharge:	8	y
Decay time after reprocessing:	100000	y

Reprocessing ratio (%)

Uranium	99.9
Plutonium	99.9
Neptunium	99.5
Americium	99.5
Curium	99.5

webKORIGEN

webKORIGEN was developed from the Oak Ridge Isotope Generation and Depletion code ORIGEN.
Starting with a given initial reactor fuel or a single target nuclide, it calculates the time evolution of nuclide densities changing due to decays and neutron-induced reactions, and determines derived nuclear properties such as masses, radioactivities, heat releases, radiotoxicities, emission of radiation, etc...

Step 1: Calculation Mode | Step 2: Reactor / Operation | Step 3: Input Summary and Run | Step 4: Display Results | Step 5: Log files

Input summary

Mode of calculation:	Reactor irradiation and decay 40 MW·d/kg
Reactor type:	BWR
Fuel:	UOX with 3.3% enrichment
Operation parameters:	No. of cycles: 5 Length of cycle: 365.24 d Load factor: 80.0 % Fuel decay time after discharge: 8 y Heavy metal mass: 20 t

Run calculation

Version 2.0.0003.0000

Display results for nuclides/elements dominant at 8 y decay

Display quantity: Mass (g)

Top Nuclides	Results
U238	1.875e+7
U235	1.270e+5
Pu239	9.244e+4
U236	9.043e+4
Xe136	5.512e+4
Pu240	5.232e+4
Xe134	3.673e+4
Nd144	3.275e+4
Ba138	3.194e+4
Ce140	3.084e+4
La139	2.954e+4
Ce142	2.760e+4
Pr141	2.742e+4
Cs133	2.702e+4
Xe132	2.527e+4
Cs137	2.432e+4
Mo100	2.188e+4
Mo98	2.080e+4
Pu241	1.962e+4
Zr96	1.949e+4

Top Elements	Results
Uranium	1.897e+7
Plutonium	1.859e+5
Xenon	1.255e+5
Neodymium	9.382e+4
Zirconium	8.758e+4
Molybdenum	8.129e+4
Cesium	6.145e+4
Cerium	5.844e+4
Barium	4.333e+4
Lanthanum	2.954e+4
Praseodymium	2.742e+4
Neptunium	9.570e+3

Totals	Results
Actinides:	1.918e+7
Fission Products:	8.191e+5
Total:	2.000e+7

Neutron and gamma rates

Neutron emission rate: 1.122E+10 n/s